The Emission and Absorption X-Ray Edges of Li*

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ABSTRACT

Measurements of the K X-ray absorption and emission edges of Li are reported. They were made with the same spectrometer at the NBS storage ring and serve to establish a 0.1 eV separation between the edges with no possibility of instrument calibration error. These results are compared with recent theories of Almbladh and Mahan describing the effects of incomplete phonon relaxation about the core hole. We conclude that these theories give a satisfactory explanation of the data.

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The Emission and Absorption X-Ray Edges of Li*

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In a recent paper, two of the present authors reported measurements of the temperature dependence of the K X-ray emission edge of Li. The results showed that the broad emission edge observed above room temperature could be resolved at liquid nitrogen temperatures into a steep high energy edge plus a peak lying about 0.7 eV below the edge.⁽¹⁾ Similar results reported earlier by Crisp and others⁽²⁾ had in recent years generally been explained as a very broad emission edge modified by self-absorption.⁽³⁾ It was shown that self absorption could not provide an explanation for the new results.

Recently C. O. Almbladh and G. D. Mahan, completed very similar calculations that give a satisfactory explanation of the experimental results.^(4,5) They treat the case where the lifetime of the core hole created by absorption is comparable to the lattice relaxation time of the lattice about the hole. Edge structure is produced because emission sometimes occurs before and sometimes after the lattice relaxation.

In this paper, we report absorption and emission measurements of the Li K edge made with the same spectrometer at the storage ring of the National Bureau of Standards (NBS). Previous measurements had not firmly established the existence of a gap between the emission and absorption edges, because the gap width is comparable to the calibration differences between the instruments used for the separate measurements. In addition we compare the emission and absorption edge shapes with the recent theoretical calculations.

A more detailed description of the experimental apparatus will be given elsewhere.⁽⁶⁾ In brief, absorption measurements were made at liquid nitrogen temperature on thin films of Li evaporated onto carbon film substrate. Emission was measured by moving a cooled emission target into the sample position and

bombarding it with 2 KV electrons. Twenty emission curves were averaged to obtain the reported results.

Both emission and absorption curves are shown in Fig. 1. The smooth curve through the emission data was obtained by broadening the sharp edged dashed curve shown with a Gaussian broadening function having a FWHM value of 0.25 eV. An identical curve fits the previously published data if it is shifted in energy by 0.04 eV. The difference is the calibration difference between the NBS and ORNL (Oak Ridge National Laboratory) spectrometers.

Fig. 1 (above) Absorption and emission spectra of Li. Dashed line is unbroadened emission edge. $\Gamma_{\frac{1}{2}}$ is FWHM value of Gaussian broadening function.

Fig. 2 (left) Derivatives of emission and absorption edges. Γ_{ph} is phonon broadening and γ is the Auger broadening of the X-ray processes.

In Fig. 2, the derivatives of the curves in Fig. 1 and of an emission curve taken at 400°C are shown along with the emission and absorption intensities calculated by G. D. Mahan for a phonon broadening FWHM value of 0.26 eV and a core hole lifetime of 30 meV. All curves have been plotted relative to the peak position representing the maximum slope of the absorption edge. The experimental curves are not strictly comparable with the theoretical curves since they represent the theoretical intensity factor folded with a slowly varying density of states. The density of states effects will change the relative magnitudes of the two peaks contributing to the emission spectra. It also probably accounts for the shoulder on the absorption curve at about ± 0.25 eV which has been attributed to a peak in the density of states above the Fermi level in Li.⁽⁷⁾

Excellent agreement between experiment and theory is obtained for the 0.1 eV separation between the peaks representing the position of maximum slope on the emission and absorption edges. Good agreement is also obtained for the relative position of the normal (-0.1 eV) and Frank-Condon shifted (-0.5 eV) peaks which contribute to the emission intensity. According to Mahan, the width of the emission edge should be about 40% greater than that of the absorption edge. The experimental data at 100°K shows FWHM values of 0.25 eV and 0.20 eV for the widths of the two edges. This corresponds to a phonon broadening of about 0.20 eV in the theories of Mahan and Almbladh. The data at 400°C has a FWHM value of 0.4 eV which corresponds to a phonon broadening of about 0.3 eV in the theory. A more complete comparison of these results to other published data will be made elsewhere. We conclude that our results are consistent with the Almbladh-Mahan theory for a core hole lifetime of 30 ± 5 meV, that phonon broadening is the major contributor to the width of the edge spectra, and that many body effects do not contribute in an important way to the explanation of the X-ray edge data in Li.

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